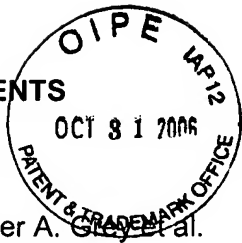


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PATENT

AF

Re: Application : Roger A. Gray et al. Case No. : 01-2619A
Serial No : 10/785,455 Unit No : 1754
Filed : 02/24/2004 Examiner : E. M. Johnson
For : Catalyst Regeneration Process

Enclosed are the following documents related to the above-identified application:

- (X) Return Receipt of Postcard
(X) Certificate of Mailing
() Declaration Under 37 C.F.R. § 1.131
() Declaration Under 37 C.F.R. § 1.132
() Assignment for Recordal
() Terminal Disclaimer 37 C.F.R. § 1.321(c)
() Information Disclosure Statement
() Notice of Appeal
Under 37 C.F.R. § 1.97(b)(1),
() Appeal Brief Under 37 C.F.R. § 1.192(d)
References, and Form PTO-1449
() Version with Markings to Show Changes
() Amendment After Allowance 37 C.F.R. § 1.312
() Issue Fee Transmittal
() Response to Restriction Requirement
() Certificate Under 37 C.F.R. § 3.73(b)
() Two-Month Extension of Time Under
() Request for Reconsideration
37 C.F.R. § 1.136 (fee noted below)
(X) Response to Examiner's Notification
() Amendment Under 37 C.F.R. § 1.121
of Non-Compliant Appeal Brief (37 CFR 41.37)

The fee has been calculated as shown below:

CLAIMS AS AMENDED

| Claims Remaining After Amendment | | Highest No. | | Add'l Fee |
|---------------------------------------|------------|------------------------|-----------------------|--------------|
| | | Previously Paid for | Present Extra Rate | |
| Total Claims: | minus : 20 | : | :x \$50 | : 0.00 |
| Ind. Claims: | minus : 3 | : | :x \$200 | : 0.00 |
| Fee for Petition of Extension of time | : | : | : | : 0.00 |
| TOTAL FEE DUE | | | \$ | : 0.00 |

- (X) No additional fee is required.
() Charge \$_____ to Deposit Account No. 01-2230. Two duplicate copies of this sheet are enclosed.
(X) Please charge any additional fees or credit overpayment to Deposit Account No. 01-2230.

I hereby certify that this correspondence is being deposited with the United States Postal Service as first-class mail, with sufficient postage, in an envelope addressed to: Mail Stop: APPEAL BRIEF-PATENTS Commissioner for Patents, P. O. Box 1450, Alexandria, VA 22313-1450 on October 26, 2006.

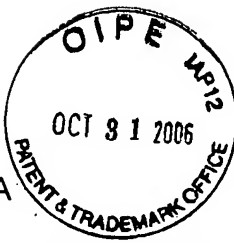
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October 26, 2006
CUSTOMER NUMBER 24114

01-2619A



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Roger A. Grey et al.

: Art Unit: 1754

Serial No.: 10/785,455

: Examiner: E.M. Johnson

Filed: 2/24/04

For: CATALYST REGENERATION PROCESS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

APPEAL BRIEF UNDER 37 C.F.R. § 1.192(d)

In response to Examiner's Notification of Non-Compliant Appeal Brief (37 CFR 41.37), mailed on October 20, 2006, please replace the previous Appeal Brief filed on August 25, 2006 with the following replacement Appeal Brief in accordance with 37 CFR 41.37(d).

I. Real Party in Interest

The real party in interest is Lyondell Chemical Technology, L.P., a subsidiary of Lyondell Chemical Company.

II. Related Appeals and Interferences

There are no other appeals or interferences known to Appellants, their representative, or assignee that will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. Status of the Claims

Claims 1-18 are pending in this application. Claims 1-18 have been rejected and are the subject of this appeal.

IV. Status of Amendments

There are no amendments filed subsequent to final rejection. Claims 1-18 on appeal are the originally filed claims.

V. Summary of Claimed Subject Matter

Appellants claim a method of regenerating a used noble metal-containing titanium or vanadium zeolite catalyst. The noble metal-containing titanium zeolite catalyst was used to catalyze the epoxidation of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer. The method comprises heating the used catalyst at a temperature of at least 250°C in the presence of a gas stream comprised of oxygen to obtain a heated product, and then reducing the heated product at a temperature of at least 20°C in the presence of a gas stream comprised of hydrogen to form a reactivated catalyst (claim 1).

Independent claim 13 also claims a method of regenerating a used noble metal-containing titanium zeolite catalyst which has been used to catalyze the epoxidation of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer. The method comprises first washing the used catalyst with a wash solvent; then heating the washed catalyst at a temperature of at least 300°C in the presence of a gas stream comprised of oxygen to obtain a heated product; and then reducing the heated product at a temperature of at least 30°C in the presence of a gas stream comprised of hydrogen to form a reactivated catalyst. Dependent claim 2 also requires the washing the used noble metal-containing titanium zeolite catalyst with a wash solvent prior heating the used catalyst.

The noble metal-containing titanium or vanadium zeolite catalyst (discussed at p. 3, l. 11 to p. 4, l. 31) is used in the epoxidation of an olefin with hydrogen and oxygen (discussed at p. 4, l. 32 to p. 6, l. 34). The regeneration procedure is discussed in detail at p. 7, l. 1 to p. 9, l. 34.

VI. Grounds of Rejection to be Reviewed on Appeal

A. Are claims 1-18, and separately claims 13-18, obvious under 35 U.S.C. § 103(a) as unpatentable over Grey (U.S. Pat. No. 6,441,204) in view of Schindler et al. (U.S. Pat. No. 6,916,756)?

VII. Argument

A. Claims 1-18 are Nonobvious over Grey in view of Schindler

1. Grey (U.S. Pat. No. 6,441,204) and Schindler (U.S. Pat. No. 6,916,756)

Grey discloses a liquid phase process for epoxidizing an olefin with hydrogen and oxygen in the presence of a catalyst mixture comprising a titanium zeolite and a supported catalyst comprising palladium on niobium-containing support (see Abstract). Grey also teaches that the epoxidation reaction can take place in the optional presence of a buffer (at Col. 4, l. 18-39).

Schindler discloses a process for regenerating a dehydrogenation catalyst. The Schindler dehydrogenation catalyst regeneration process comprises (a) flushing with inert gas; (b) passing an oxygen-containing gas mixture; (c) optionally passing an oxygen-containing gas mixture at a different pressure and gas hourly velocity than step (b); (d) optionally changing the pressure repeatedly, rapidly and in opposite directions; (e) flushing with an inert gas; (f) activating the catalyst by means of hydrogen; where at least one of the steps (c) or (d) is carried out and the entire regeneration process is carried out at from 300 to 800°C (see Abstract).

The dehydrogenation catalyst of Schindler comprises one or more group VIII transition metals (preferably Pt or Pd) on a support. The support is "a metal oxide selected from the group consisting of zirconium dioxide, zinc oxide, aluminum oxide, silicon dioxide, titanium dioxide, magnesium oxide, lanthanum oxide, cerium oxide and mixtures thereof as support. Preferred supports are zirconium dioxide and/or silicon dioxide; particular preference is given to mixtures of zirconium dioxide and silicon dioxide" (see Col. 4, l. 3-9).

2. The PTO Position: Appellants' Claimed Process is Obvious over Grey in view of Schindler

The Examiner's rationale for rejecting claims 1-18 is located in the final Office Action dated January 25, 2006. The Examiner states that "It would have been obvious to one of ordinary skill in the art at the time to modify the teachings of Grey, by first treating a used catalyst comprised of titanium, silicon, and a noble metal, such as palladium, with an oxygen gas and then a secondary treatment with hydrogen gas at a temperature of at least 250 C and 20 C respectively, because Schindler et al. continues to teach wherein a catalyst comprised of mixtures of titanium oxides, as well as silicon oxides along with a noble metal, such as palladium may be regenerated by passing an oxygen-containing gas through the catalyst, and then treating the catalyst with a hydrogen gas, while the entire regeneration process is carried out at a temperature from 300-800 C. Such modification would have been obvious to one of ordinary skill in the art, because one of ordinary skill in the art would have expected a process for treating a catalyst comprised of a titanium zeolite with a noble metal, as taught by Schindler et al. to have been similarly useful and applicable to a process for using a catalyst comprised of a titanium zeolite and a supported catalyst comprising palladium as taught by Grey." See pages 3-4 of the January 25, 2006 Office Action.

3. The Cited Prior Art Does Not Suggest to One of Ordinary Skill in the Art That They Should Make Appellants' Claimed Process

"A proper analysis under § 103 requires, inter alia, consideration of two factors: (1) whether the prior art would have suggested to those of ordinary skill in the art that they should make the claimed composition or device, or carry out the claimed process; and (2) whether the prior art would also have revealed that in so making or carrying out, those of ordinary skill would have a reasonable expectation of success" (emphasis added). *In re Vaeck*, 947 F.2d 488 (Fed. Cir. 1991).

Examiner has failed to make a prima facie case of obviousness since the § 103 analysis fails under both factors described above. First, the combination of Grey and Schindler would not suggest to one of ordinary skill in the art that they should make Appellants' claimed regeneration method since neither reference discloses the required

noble metal-containing titanium zeolite catalyst. The § 103 analysis also fails the second factor. Grey and Schindler do not reveal to those of ordinary skill a reasonable expectation of success because Grey does not even teach that its catalyst mixtures require regeneration following use in an epoxidation reaction.

a. Neither Grey Nor Schindler Teach Appellants' Required Noble Metal-Containing Titanium or Vanadium Zeolite Catalyst

Appellants claim a process to regenerate a noble metal-containing titanium or vanadium zeolite catalyst. The plain meaning of the term "noble metal-containing titanium zeolite catalyst" is obvious to one skilled in the art. A "noble metal-containing titanium zeolite catalyst" is a catalyst in which the titanium zeolite contains a noble metal. The current application clearly describes that the noble metal-containing titanium zeolite catalyst is one in which the noble metal is ***incorporated into the zeolite*** by impregnation, ion exchange, or the like (see the present application at page 4, l. 4-8).

Although Grey discloses a liquid phase epoxidation process similar to Appellants' epoxidation process, Grey does not disclose the "noble metal-containing titanium zeolite catalysts" required by Appellants' current invention. Rather, Grey teaches a catalyst mixture that comprises two separate components: (a) a titanium zeolite; and (b) a palladium on niobium-containing support (e.g., Pd/Nb₂O₅). Thus, Grey does not teach a titanium zeolite that contains a noble metal as required by Appellants, but rather teaches a mixture of two separate components that is different than Appellants' required noble metal-containing titanium zeolite catalyst.

Schindler also does not disclose Appellants' required "noble metal-containing titanium zeolite catalyst." Schindler does teach noble metal-containing catalysts in which a noble metal is supported on a support. However, the support taught by Schindler is not a zeolite. Rather, Schindler only discloses ***amorphous (i.e., non-zeolite)*** metal oxides. Specifically, Schindler states that the "dehydrogenation catalysts which can be regenerated according to the present invention generally comprise a support and an active composition. The support comprises a heat-resistant oxide or mixed oxide. The dehydrogenation catalyst preferably comprises a metal oxide

selected from the group consisting of zirconium dioxide, zinc oxide, aluminum oxide, silicon dioxide, titanium dioxide, magnesium oxide, lanthanum oxide, cerium oxide and mixtures thereof as support. Preferred supports are zirconium dioxide and/or silicon dioxide; particular preference is given to mixtures of zirconium dioxide and silicon dioxide" (at Col. 3., l. 66 to Col. 4, l. 9).

Thus, the Schindler dehydrogenation catalysts do not contain titanium zeolites, such as titanium silicalites (e.g., TS-1), as required by Appellants. Instead, the Schindler dehydrogenation catalyst contains non-zeolitic metal oxides. When mixed oxides are taught, specifically for zirconia-silica mixed oxides, these mixed oxides are taught as being prepared by simply mixing silica and zirconia (see Col. 4, l. 41-62).

Schindler does not teach zeolite supports which are produced by reacting a titanium compound, a silicon source, and a templating agent under hydrothermal conditions to produce a molecular sieve. The closest that Schindler comes to teaching the use of zeolites is found at Col. 4, l. 66 to Col. 5, l. 19. In this section, Schindler discloses dehydrogenation catalysts having "a defined pore structure" and produced by "addition of various polymers during production of the catalyst." However, these mixed oxide catalysts are taught to have "macropores in the range from 200 to 5000 nm" (i.e., 2000-50,000 Å). These macroporous, amorphous mixed oxides are not zeolites. In contrast, titanium zeolites are microporous, crystalline molecular sieves having micropores of less than 20 Å in pore diameter (versus the 2000-50,000 Å macropores of Schindler's amorphous mixed oxides). Thus, Schindler at best teaches only the supported noble metal component of Grey without the titanium zeolite component.

The following table, which summarizes the Examples in the current application and the cited references, clearly demonstrates the differences between Appellants' required catalyst and those taught in Grey and Schindler.

Table 1: Difference between Currently Required Catalyst and Prior Art Catalysts, as seen in Examples

| Current Application | Grey | Schindler |
|---------------------|--|---------------------------------------|
| Pd/TS-1 | TS-1 + Pd/Nb ₂ O ₅ | Pt/ZrO ₂ .SiO ₂ |

Since Grey and Schindler do not teach Appellants' required "noble metal-containing titanium zeolite catalysts," there is nothing in a combination of these references that suggests a process for regenerating noble metal-containing titanium zeolite catalysts.

b. Grey Does Not Teach that Its Catalyst Mixtures Require Regeneration

The mere fact that references can be combined or modified does not render the resultant combination obvious, unless the prior art also suggests the desirability of the combination. In this case, there is no suggestion or incentive to combine the cited prior art because Grey does not teach that its catalyst mixtures require regeneration.

As described above, Grey does not teach noble metal-containing titanium or vanadium zeolite catalysts but rather teaches a catalyst mixture comprising a titanium zeolite and a palladium on niobium-containing support (e.g., TS-1 + Pd/Nb₂O₅). In addition, Grey does not even disclose that its catalyst mixtures require regeneration. Therefore, Grey does not provide the motivation for one of ordinary skill in the art to combine Grey with the teachings of Schindler.

Schindler discloses that its **dehydrogenation** catalysts require regeneration. However, although Schindler teaches that its Pt/ZrO₂.SiO₂ catalysts deactivate under the high temperature (605-625°C – see Examples) dehydrogenation conditions, there is nothing in Schindler that would suggest that its Pt/ZrO₂.SiO₂ catalysts would be detrimentally impacted and require regeneration following the **low temperature** (60°C –

see Example 2) epoxidation reaction of Grey employing a catalyst mixture of a titanium zeolite and $\text{Pt/ZrO}_2\cdot\text{SiO}_2$.

Thus, because neither Grey nor Schindler teach that a catalyst mixture of a titanium zeolite and a supported noble metal (i.e., TS-1 + $\text{Pd/Nb}_2\text{O}_5$) is deactivated in the epoxidation reaction of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer, one of ordinary skill in the art would have no motivation to combine Schindler's regeneration teachings with the epoxidation reaction disclosure of Grey. This is, of course, in addition to the fact that neither offers motivation to regenerate Appellants' required noble metal-containing titanium or vanadium zeolite catalysts since neither Grey nor Schindler teach the presently required catalyst (as described above).

The § 103 analysis thus fails under both factors described above. Grey and Schindler would not suggest to one of ordinary skill in the art that they should make Appellants' claimed regeneration process for a noble metal-containing titanium zeolite catalyst that was used to catalyze the epoxidation of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer. Grey does not disclose Appellants' required catalyst, but rather teaches catalyst mixtures. Grey does not even disclose that its catalyst mixtures even deactivate or require regeneration, nor does Grey teach any regeneration process. Thus, there is nothing to suggest to one of ordinary skill in the art that they should combine the regeneration process of Schindler (for a dehydrogenation catalyst that is different than Appellants' required zeolitic-based catalyst) with the epoxidation process of Grey.

4. Claims 2-3 and 13-18 are Nonobvious over Grey in view of Schlinder

The current claims 2-3 and 13-18 require washing the used noble metal-containing titanium zeolite catalyst with a wash solvent prior heating the used catalyst.

As discussed above, Grey discloses an epoxidation procedure using a catalyst mixture (TS-1 + $\text{Pd/Nb}_2\text{O}_5$) but does not disclose a regeneration procedure or even that its catalyst mixture requires regeneration. Schindler discloses a regeneration procedure for a dehydrogenation catalyst ($\text{Pt/ZrO}_2\cdot\text{SiO}_2$) that comprises (a) flushing with inert gas;

(b) passing an oxygen-containing gas mixture; (c) optionally passing an oxygen-containing gas mixture at a different pressure and gas hourly velocity than step (b); (d) optionally changing the pressure repeatedly, rapidly and in opposite directions; (e) flushing with an inert gas; (f) activating the catalyst by means of hydrogen; where at least one of the steps (c) or (d) is carried out and the entire regeneration process is carried out at from 300 to 800°C. Schindler does not disclose first washing its Pt/ZrO₂.SiO₂ catalyst with a wash solvent prior to heating the used catalyst. In fact, Schindler does not mention the use of any solvent other than for the preparation of its catalyst.

Appellants' own results unexpectedly indicate that washing prior to calcination and reduction results in substantially increased productivity compared to a used catalyst and a used catalyst that is not washed prior to calcination and reduction, with essentially the same PO/POE selectivity. The results also indicate substantially improved propane selectivity for the regenerated catalysts. Lower propane selectivity indicates lower production of unwanted byproduct propane produced by the hydrogenation of propylene by hydrogen. Table 2 below summarizes the results from the current application's Table 1, detailing the same used catalyst (2B) that undergoes calcination and reduction (3B) versus washing, calcination and reduction (4B).

Table 2: Comparison of Epoxidation Activity Following Regeneration Procedures

| Catalyst | Treatment | Productivity | PO/POE Selectivity (%) | Propane Selectivity (%) |
|----------|-------------------------------|--------------|------------------------|-------------------------|
| 2B * | Recovered and dried | 0.16 | 94 | 51 |
| 3B | Calcined and reduced | 0.22 | 92 | 27 |
| 6B | Washed, calcined, and reduced | 0.3 | 93 | 19 |

There is nothing in Grey or Schindler that would suggest the unexpectedly improved productivity and propane selectivity resulting from performing a washing step prior to heating and reducing the used catalyst.

In sum, there is nothing in a combination of Grey and Schindler that would have suggested to one of ordinary skill in the art that they should wash the used noble metal-containing titanium zeolite catalyst with a wash solvent prior heating and reducing the used catalyst, or that would have revealed to those of ordinary skill a reasonable expectation of success in carrying out such a regeneration.

B. Summary

In sum, Grey and Schindler fail to teach or suggest a method of regenerating a used noble metal-containing titanium or vanadium zeolite catalyst, that has been used to catalyze the epoxidation of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer, which comprises heating the used catalyst at a temperature of at least 250°C in the presence of a gas stream comprised of oxygen to obtain a heated product, and then reducing the heated product at a temperature of at least 20°C in the presence of a gas stream comprised of hydrogen to form a reactivated catalyst (claim 1). Nor do Grey and Schindler disclose first washing the used noble metal-containing titanium zeolite catalyst with a wash solvent prior heating and reducing the used catalyst. Because the cited prior art does not suggest Appellants' claimed process, the claimed process meets the patentability requirements of Section 103(a).

Because the differences between the claimed process and those disclosed by Grey and Schindler would not have been obvious to a skilled person in this field, the Board should reverse the Section 103 rejection.

VIII. Conclusion

Appellants respectfully ask the Board of Patent Appeals and Interferences to reverse the Examiner's rejections of claims 1-18 under 35 U.S.C. § 103(a).

I hereby certify that this correspondence is being deposited with the United States Postal Service as first-class mail, with sufficient postage, in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on October 26, 2006.

Kevin M. Carroll
Name of person signing

Kevin M. Carroll
Signature

Respectfully submitted,

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Claims Appendix

1. (original) A method of regenerating a used noble metal-containing titanium zeolite catalyst comprising the steps of:

- (a) heating the used catalyst at a temperature of at least 250°C in the presence of a gas stream comprised of oxygen to obtain a heated product; and
- (b) reducing the heated product at a temperature of at least 20°C in the presence of a gas stream comprised of hydrogen to form a reactivated catalyst;

wherein the noble metal-containing titanium zeolite catalyst was used to catalyze the epoxidation of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer.

2. (original) The method of claim 1 which comprises washing the used noble metal-containing titanium zeolite catalyst with a wash solvent prior to step (a).

3. (original) The method of claim 2 wherein the wash solvent is selected from the group consisting of water, aliphatic alcohols, and mixtures thereof.

4. (original) The method of claim 1 wherein the used catalyst is heated at a temperature greater than about 300°C.

5. (original) The method of claim 1 wherein the gas stream comprised of oxygen is air.

6. (original) The method of claim 1 wherein reduction step (b) is performed at a temperature of at least 30°C.

7. (original) The method of claim 1 wherein the used noble metal-containing titanium zeolite catalyst comprises titanium silicalite and palladium.

8. (original) The method of claim 1 wherein the used noble metal-containing titanium zeolite catalyst comprises titanium silicalite, palladium, and one or more metals selected from the group consisting of gold and platinum.

9. (original) The method of claim 1 wherein the used noble metal-containing titanium zeolite catalyst comprises a palladium-containing titanium zeolite and a palladium-free titanium zeolite.

10. (original) The method of claim **1** which comprises heating the used catalyst at a temperature of at least 250°C in the absence of oxygen prior to step (a).

11. (original) The method of claim **1** wherein the reaction solvent is selected from the group consisting of water, C₁-C₄ alcohols, supercritical CO₂, and mixtures thereof.

12. (original) The method of claim **1** wherein the buffer comprises an anion and a cation, wherein the anion is selected from the group consisting of phosphate, carbonate, bicarbonate, carboxylate, citrate, borate, hydroxide, silicate, aluminosilicate, and mixtures thereof, and the cation is selected from the group consisting of ammonium, alkylammonium, alkali metal, alkaline earth metal, and mixtures thereof.

13. (original) A method of regenerating a used noble metal-containing titanium zeolite catalyst comprising the steps of:

- (a) washing the used catalyst with a wash solvent;
- (b) heating the washed catalyst at a temperature of at least 300°C in the presence of a gas stream comprised of oxygen to obtain a heated product; and
- (c) reducing the heated product of step (b) at a temperature of at least 30°C in the presence of a gas stream comprised of hydrogen to form a reactivated catalyst;

wherein the used noble metal-containing titanium zeolite catalyst was used to catalyze the epoxidation of an olefin with hydrogen and oxygen in the presence of at least one reaction solvent and at least one buffer.

14. (original) The method of claim **13** wherein the wash solvent is selected from the group consisting of water, aliphatic alcohols, and mixtures thereof.

15. (original) The method of claim **13** which comprises heating the washed catalyst at a temperature of at least 300°C in the absence of oxygen prior to step (b).

16. (original) The method of claim **13** wherein the used noble metal-containing titanium zeolite catalyst comprises titanium silicalite and palladium.

17. (original) The method of claim **13** wherein the used noble metal-containing titanium zeolite catalyst comprises titanium silicalite, palladium, and one or more metals selected from the group consisting of gold and platinum.

18. (original) The method of claim **13** wherein the used noble metal-containing titanium zeolite catalyst comprises a palladium-containing titanium zeolite and a palladium-free titanium zeolite.

Evidence Appendix

None

Related Proceedings Appendix

None